

(12) PATENT ABRIDGEMENT (11) Document No. AU-B-20822/83
 (19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 568608

(51)4 International Patent Classification

C08F 283/00 B32B 017/10 C03C 027/10 C08J 005/12

(21) Application No. : 20822/83 (22) Application Date : 31.10.83

(30) Priority Data

(31) Number (32) Date (33) Country
 8231737 05.11.82 GB UNITED KINGDOM

(43) Publication Date : 10.05.84

(44) Publication Date of Accepted Application : 07.01.88

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(54) Title
 RADIATION CURABLE URETHANE ACRYLATE COMPOSITIONS

(56) Prior Art Documents
 EP 10355
 50710/79 536517

(57) Claim

1. A radiation-curable fluid adhesive composition for the production of clear glass laminate and of the kind comprising (a) a urethane acrylate composition in (b) a liquid $\alpha\beta$ -ethylenically unsaturated diluent component comprising acrylic acid and a monoester of acrylic acid, said composition also optionally containing (c) a photoinitiator, characterised in that said urethane acrylate component comprises at least one urethane acrylate of the kind obtainable by reaction of an oligomeric urethane intermediate having end groups selected from -OH and -NCO with an $\alpha\beta$ -ethylenically unsaturated compound which has a group reactive with an end group of the intermediate and also a carboxy (-COO) group attached to a carbon atom which is attached to a methylene group by a double bond, more than 50% by weight of said urethane acrylate component is material having a molecular weight (M_n) above 1500, and said urethane acrylate component forms from 45% to 75% by weight of the composition, and said diluent component comprises (i) acrylic acid, (ii) an effective amount of at least one acrylate selected from monoesters of acrylic acid and alkanols having from 1 to 6 carbon atoms and substituted derivatives of such alkanols, wherein each substituent contains only carbon, hydrogen and oxygen atoms, and, optionally, (iii) a multiacrylate; said component (i) being present in an amount of from 35% to 90% by weight,

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COMPLETE SPECIFICATION

(ORIGINAL)

FOR OFFICE USE :

568608

Class

Int. Cl

Application Number : 26822/83
Lodged :Complete Specification Lodged :
Accepted :
Published :

Priority :

This document contains the
amendments made under
Section 49.

and is correct for printing

Related Art :

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Complete Specification for the invention entitled :

"RADIATION CURABLE ADHESIVE COMPOSITIONS"

The following statement is a full description of this invention, including the best method of performing
it known to me/us :

RADIATION-CURABLE ADHESIVE COMPOSITIONS

This invention relates to radiation-curable compositions suitable for use as adhesives for bonding glass, and in particular for the manufacture of clear glass laminates.

There have been many proposals in the literature for such compositions. In general they comprise a viscous or solid radiation-curable ethylenically unsaturated macromolecular or resinous, e.g. oligomer or polymer component dissolved or dispersed in a liquid diluent comprising one or more ethylenically unsaturated monomers which are capable of copolymerising with the said component.

To be commercially acceptable as an adhesive for bonding glass in large scale manufacturing operations such as the manufacture of laminated glass products, the compositions must not only bond adequately in the cured state to the glass and to the other substrate, which may or may not be glass, but must also have an adequate shelf life and yet be able to be cured rapidly when exposed to the chosen irradiation, and must be available in a form having a viscosity sufficiently low in the uncured state to permit its facile application to a substrate in a thin and uniform layer without the need for sophisticated equipment.

For the production of laminated glass products intended to be used as windows or doors or in vehicles, the cured product from the composition must also be clear, colourless and transparent, preferably have a refractive index at least close to that of glass, exhibit adequate water- or moisture-resistance

the product, which will usually be oligomeric, is called, for ease of reference, a urethane acrylate.

10 One class of composition that has shown particular promise as an adhesive for glass contains a urethane acrylate as the resinous component and a reactive diluent such as acrylic acid, e.g. as described in European patent publication 10355. However, an improvement in the bond obtained between glass and plastics materials, especially vinyl chloride polymers, using the composition would be desirable. In the production of glass laminates, two properties of the bond which are important are the adhesive or bond strength, e.g. as measured in a Peel Test, and the performance of the bond under impact. The latter can be gauged from the performance of the bond in the Peel Test. If the force required when performing the Peel Test at a steady parting rate of e.g. 30 cm/minute is not uniform and/or if it drops markedly if the parting rate is increased sharply, e.g. as when the components are pulled apart sharply by hand, the bond is said to exhibit "driness" and is less likely to perform well under impact; in particular, delamination and/or splintering of the glass may occur. 20 It is in these properties of bond strength and driness in particular where improvement is desired; i.e. an increase in bond strength and/or a reduction in "driness" which can alternatively be described as an increase in the "softness" of the bond. Of course, such improvement should be obtained without unacceptable deterioration of other properties,

and/or a reduction in "driness".

The acrylate component of the diluent may be an acrylate of an alkanol having 1 to 6 carbon atoms or a substituted derivative of such an alkanol, which substituent preferably contains only carbon, hydrogen and oxygen atoms e.g. as in alkyl-, alkoxyalkyl-, epoxyalkyl and hydroxyalkyl-acrylates. A mixture of such acrylates may also be used.

Examples of suitable acrylates are ethyl acrylate, n-butyl acrylate, glycidyl acrylate, 2-ethoxyethyl acrylate and 2-hydroxyethyl acrylate. Because of the volatility of the lower alkyl acrylates, it is preferred that the alkanol have at least 3 carbon atoms. Alkanols having over 6 carbon atoms, on the other hand, do not appear to give the desired improvement in the bond. Preferably the alkanol will have the structure



wherein R is -H, -OH or a monovalent organic group containing atoms selected only from carbon, oxygen and hydrogen, and having not more than 4 atoms in a chain attached to the free valency, and each R' is individually selected from -H and -CH₃, or R and R' together may form a divalent group containing atoms selected only from carbon, oxygen and hydrogen, the sum of the carbon and oxygen atoms in said alcohol being from 4 to 7.

n-Butyl acrylate is particularly preferred because compositions can be formed therefrom which have not only good bond strength and low driness but also excellent water- and

production of transparent laminates, not all of the specified acrylates may be suitable for use with all urethane acrylate compositions; some combinations tend to give rise to a lack of clarity in the cured material. In some cases where turbidity has been observed in the uncured composition, this has disappeared in the cured material but whether a particular combination of urethane acrylate and monoethyl acrylate is suitable can readily be determined by simple experiment.

10
The urethane acrylate component, may comprise one or a mixture of urethane acrylates which are preferably oligomeric. The nature of the urethane acrylate component and its concentration in the composition are important factors in the reactivity of the composition, the adhesive strength of the bond formed therefrom and the flexibility of a film of the cured composition. This last property is important where the composition is to be used to form a laminate from materials having different thermal expansion coefficients and also affects the flexural strength and impact resistance of laminates. Increasing the concentration generally leads to an improvement in reactivity and adhesive strength but also increases the viscosity thereby making the composition less easy to apply. Preferably the urethane acrylate will form about 45 to about 75% by weight of the composition, more preferably about 50 to about 70%.

20

Urethane acrylates are notionally obtainable by the reaction of a polyol and a polyisocyanate to form an oligomeric

2000-3000, e.g. at least 4000. The term molecular weight as used herein refers to number average molecular weight (M_N) expressed in terms of polystyrene equivalent as measured by, for example, Gas Phase Chromatography (GPC).

The processes most usually employed to produce the urethane acrylates generally yield mixtures of oligomers, and these mixtures may also include in minor amounts monomeric reaction by-products such as the reaction products of the polyisocyanate or the polyol with the ω -ethylenically unsaturated compound. These mixtures or fractions thereof may be used as such in the compositions of the present invention.

In general it is desired that the urethane acrylate component comprise at least mainly, and preferably largely, material having a functionality, meaning number of ω -ethylenically unsaturated groups per molecule, of at least about 2. Usually it will be preferred to have on average about two, e.g. from about 1.5 to about 2.5, preferably about 1.7 to about 2.3, such groups per molecule.

The chemical nature of the urethane acrylate component also affects the properties of the composition and particularly the resistance to water and to u.v. irradiation of the adhesive layer obtained from the composition in a laminate. We have found that the better results are obtained where the urethane acrylate is derived at least mainly from polyol which contains ether groups, e.g. as in polyether polyols and polyetherester polyols.

Best results have been obtained where at least the major part of the urethane acrylate component comprises urethane acrylate derived from polyol which is at least mainly polyether polyol e.g. a polymer of ethylene oxide and/or propylene oxide, and which preferably is free or substantially free of ester groups.

10
The polyisocyanates may be aromatic, e.g. as in phenylene diisocyanates, toluene diisocyanates and bis(isocyanatoaryl)-alkanes. However, it is preferred that most and preferably all or substantially all of the isocyanate groups of the polyisocyanate are attached to aliphatic carbon atoms, e.g. as in polymethylene, e.g. hexamethylene diisocyanates, bis(isocyanatomethyl)cyclohexanes, bis(isocyanatocycloalkyl)-alkanes e.g. bis(isocyanatocyclohexyl)methanes and isophorone diisocyanate.

It is preferred that all or substantially all of the unsaturation in the urethane acrylate component of the composition is terminal $\alpha\beta$ -unsaturation.

20
It has been observed that an improvement in the "softness" (i.e. reduction in the "driness") of the bond can be obtained, especially where the amount of acrylic acid exceeds the amount of acrylate in the diluent, if the urethane acrylate comprises a mixture having, preferably as the main component, (a) generally difunctional urethane acrylate material which preferably contains ether groups in the oligomer or polymer chains and which also

Component (b) is effective in even quite small amounts, e.g. when it forms 1% or even less of the total urethane acrylate component. As the amount of this component in the urethane acrylate is raised, the improvement gained appears to continue to increase to a maximum but thereafter commences to decrease. Also, the presence of large amounts appears to have an adverse effect on other properties such as water resistance. It is therefore preferred that component (b) forms no more than a minor amount of the urethane acrylate material. Best overall results are generally obtained when component (b) forms from about 0.5% to about 30%, preferably from about 0.8% to about 20%, more preferably from about 1% to 15% and most preferably from about 3% to about 15% of the urethane acrylate, by weight.

The composition may, if desired, contain other materials in addition to the urethane acrylate, acrylic acid and specified acrylate, and these may be polymerisable or non-polymerisable.

For example, a resinous or polymeric material in addition to the urethane acrylate may also be present. Such material may be unsaturated, e.g. as in $\alpha\beta$ -ethylenically unsaturated polyesters, but preferably will be at least substantially saturated e.g. as in epoxy resins, especially those derived from epichlorhydrin and bis-phenols such as bis-phenol A. Such additional resinous material will not normally form more than 50% by weight of the total composition, however.

The inclusion of balsamic resins is particularly preferred

monomeric urethane acrylates and esters of acrylic acid with alkanols having 8 or more carbon atoms; methacrylates; multiacrylates and other $\alpha\beta$ -ethylenically unsaturated compounds. However, in general it is found that the better overall combinations of properties of the composition, especially in respect of bond strength, water- and u.v.-resistance, and rate of cure, are obtained as the total combined amount of acrylic acid and specified acrylate in the diluent is increased. It is thus preferred that at least about 50% by weight of the $\alpha\beta$ -ethylenically unsaturated acid content of the diluent is acrylic acid and that the acrylic acid and specified acrylate together form at least about 50% by weight of the diluent and preferably substantially more.

The presence of multiacrylates (i.e. the di- or higher esters of acrylic acid with polyhydric alcohols such as neopentylglycol diacrylate, trimethylolpropane triacrylate and pentaerythritol tri- and tetra-acrylate), can also be tolerated. However, the bond strength of the composition appears to increase with decrease in the amount of multiacrylate present and while amounts thereof up to 40% by weight of the diluent can be accepted, smaller amounts are preferred e.g. not more than 30% and more preferably not more than 20% of the diluent, by weight.

It has further been found that the inclusion of $\alpha\beta$ -ethylenically unsaturated compounds capable of forming salts with acrylic acid e.g. N,N-dialkyl amino alkyl esters, can lead

has been absorbed by the adhesive layer. It therefore depends on the spectral distribution of the source, the power thereof, the distance thereof from the substrate to be irradiated and the optical transmission of that layer of the substrate which must be penetrated by the light before reaching the adhesive itself. Thus, glass and synthetic resins all have some significant extinction coefficient in the u.v. range and, consequently, the irradiation duration must be adapted to the optical properties of each material used.

10
As u.v. irradiation sources, any having an emission spectrum largely comprised above 0.3μ is convenient, e.g. mercury vapor lamps. One or more lamps of 20 W to about 10 KW can be used, e.g. a 2 kW lamp of type HTQ7 made by PHILIPS or a high-pressure mercury vapor lamp giving 80 W/cm made by HANOVIA. Argon or krypton lamps can also be used.

Preferably, the polymerisation is carried out only by irradiation with no further heat than that resulting from the light source. In general, it is not necessary that such heat be removed, e.g. by cooling. In general, cure can be completed within a few seconds.

20
The thickness of the adhesive layer, expressed in grams per unit surface area, can be between about 5 and about 100 g/m^2 or even exceed such limit. Best adhesion results, especially when the laminate is subjected to long exposures to moisture, are obtained when using about 15 to about 60 g/m^2 of

which is selected from glass sheets and clear plastics sheets using an adhesive, and wherein as adhesive there is used the composition in accordance with the present invention and the assembly of sheets with a layer of said adhesive composition therebetween is exposed to irradiation to cure the adhesive and bond the sheets together.

10 A particularly important feature of the invention is that it enables the replacement of the polyvinylbutyral conventionally employed as an interlayer in safety glass laminates by alternative plastics materials which perform better at elevated temperatures, especially at temperatures of 40°C or more at which polyvinyl butyral tends to lose much of its strength. A particular example is vinyl chloride polymer e.g. polyvinyl chloride.

20 Thus, in accordance with yet another embodiment of the invention, a method is provided for producing a clear glass laminate in which one face of a clear plastics foil or film interlayer is bonded to a glass sheet and the other face is bonded to a second sheet which is selected from glass sheets and clear plastics sheets, wherein the bonding of at least said glass sheet to said interlayer is effected by means of an adhesive composition in accordance with the present invention and the assembly of glass sheet and interlayer with a layer of said adhesive composition therebetween is exposed to irradiation to cure the adhesive and bond the sheet and interlayer together.

The affected area is characterised by the presence of one or more of the following: bubbling and/or blisters, striations and the presence of interference colours.

U.V. Resistance: To each of the two long edges of one face of a 50 x 25 x 4 mm glass plate is fixed a 1-2 mm wide spacer strip 1 mm thick. An excess of the composition is disposed between the spacers and a second 50 x 25 x 4 mm glass plate is pressed on top. After curing the composition, the whole assembly is exposed to irradiation at 50 - 65°C from a Hanau 160 watt u.v. lamp placed 25 cm from the sample. The u.v. resistance is estimated from the degree to which the adhesive layer has become discoloured after 63 hours exposure and is recorded on the scale 0 - 5 where 0 means there has been no observable colour change, 5 means a deep yellow colour has developed and numerals 4 to 1 relate to progressively lighter yellow colours.

Bond strength was measured by a Peel Test according to ASTM D 1876-69. A 4 x 40 x 100 mm glass plate is bonded to a 40 x 165 mm sample of pvc film, e.g. Storey's VIBAK VB 24, using a sample of the composition. The pvc film is then cut to produce a centrally disposed 25 mm wide band and the lateral bands are removed to avoid any edge effects. The central pvc band is then peeled from the glass strip at 180° and a parting speed of 30 cm/minute using an Instron or similar machine. The resistance, measured in g/cm is a measure of the bond strength.

Driness. The level of "driness" of the bond is determined by

bond well to a variety of plastics materials and have a refractive index close or equal to that of glass.

Example 1

Adhesive compositions A, B and C were prepared having the following compositions and tested for bond strength, water resistance and u.v. resistance. The results are shown in Table 1.

	A (Comparative)	B	C
Urethane acrylate	58.0	58.0	58.0
acrylic acid	38.8	29.1	19.4
n-butyl acrylate	nil	9.7	19.4
DMAEMA*	1.6	1.6	1.6
benzophenone	1.6	1.6	1.6

*DMAEMA is N,N-dimethylaminoethyl methacrylate

The urethane acrylate was a low melting point solid containing 0.4 - 0.5 eq/kg unsaturation and marketed as Oligomer AJ 17 by SNPE, France. Analysis showed it to contain residues of ethylene glycol, propylene-1,2-glycol, adipic acid, acrylic acid and toluene-2,4-diisocyanate (TDI), and to have a molecular weight M_N of about 5000-5500.

The relatively low values recorded for bond strength, as compared with those recorded for Composition C, are believed to be due, at least in part, to the low molecular weight of the urethane acrylate.

Example 3

Compositions J, K and L, corresponding to A, B and C of Example 1 were prepared but using as the urethane acrylate a material having a molecular weight of 5000, and containing on average 2 α,β -ethylenically unsaturated groups per molecule and available as Ebecryl 230 from UCB, Belgium. On analysis, this material was found to contain residues from poly(oxypropylene)-glycol, hexamethylene diisocyanate (HMDI) and acrylic acid. The results were as follows:

<u>Composition</u>	<u>bond strength (g/cm)</u>	<u>u.v. resistance</u>	<u>water resistance</u>
J (Comparative)	140-180	0	2-3
K	480	0	4
L	650	0	4-5

Example 5

As a test of the suitability of the compositions in the production of safety laminates, glass/pvc/glass laminates formed using as adhesive layers compositions N, P, R and S were subjected within a few hours of preparation to a falling ball test as follows. A 2.06 Kg ball was dropped repeatedly from a height of 3 m on to a face of a 50.6 cm x 50.6 cm laminate made using the adhesive and comprising a 0.76 mm film of "Vibak" VB 24 (a pvc film marketed by Storey Brothers of U.K.) between two 4 mm glass sheets.

In each case, it required 6-8 impacts before the ball passed through the laminate and detailed comments are as follows.

<u>Composition</u>	<u>Comments</u>
N	No sign of driness but a small amount of glass on opposite side from impact falls away on 1st Impact. More obvious on second impact.
P	As N but less glass falls away.
R	Some signs of delamination appear during test and more glass falls away after impact than in P.
S	Less satisfactory than R. Delamination appears along line of fracture on 1st impact.

The average number of strikes required before the ball passed completely through the laminate was 7-8 and in some samples the number of strikes exceeded 10, reflecting very high impact resistance.

It was also observed that when the glass shattered, the fragments and splinters largely remained firmly adhered to the interlayer and little, if any, glass separated.

When the test was repeated with the laminate heated to 40°C, the average number of strikes required was about 5-6.

Similar results were obtained in two further tests when the amount of additive was altered from 2 parts to 5 parts and 10 parts, respectively.

By way of comparison, the same test was also applied to samples of commercially available laminated glass and the results are tabulated below.

<u>Type of Laminated Glass</u>	<u>Average number of strikes required before ball passed completely through laminate</u>
A. 4 mm glass/0.76 mm interlayer/ 4 mm glass, sold commercially as "KINON"	5
B. 4 mm glass/0.76 mm interlayer/ 4 mm glass, sold commercially as "SIV"	4

Examples 8 and 9

The following Examples illustrate the inclusion of a polyester containing terminal unsaturation in the oligomeric component.

Example	8	9
Ebecryl 230	50	50
Polyester	30	30
AA	13	13
n-Bu A	0	5
HEA	5	0
Benzophenone	2	2
water resistance	3	3
adhesion (g/cm)	320	180

AA is acrylic acid

n-Bu A is n-butyl acrylate

HEA is hydroxyethyl acrylate

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Example	10 (Comparative)	11	12	13	14	15	16 (Comparative)
Acrylic Acid (parts)	38.5	33	27.5	18.5	14	9.5	0
n-Butyl Acrylate (parts)	0	5.5	11.0	20	24.5	29.0	38.5
Water Resistance	<2	3.5	4.5-5	4.5-5	4	2.5	n.m.
Cure Time (secs)	30	n.m.	n.m.	40-50	n.m.	40-50	n.m.
Bond Strength (g/cm)	1300	1600	2400	2000	600	400	negligable

n.m. means "not measured"

Comments on the performance in the Peel Test of bonds formed using the above compositions:

10 Example 10: Difficult to record bond strength because measurements vary substantially from test to test and during each test. Bond strength drops sharply if parting is rapid.

Example 11: Similar to 10.

Example 12: Similar to 10.

Example 13: Parting force fairly uniform at parting rate of 30 cm⁻¹ sec. Some drop in bond strength if parted by sharp manual movement.

Example 14: Parting force constant at 30 cm sec⁻¹ and no drop in bond strength if parted sharply.

Example 15: As 14 but bond strength barely adequate.

<u>Example No.</u>	24	25	26	27
Nature of multiacrylate	PETA	PETEA*	PETEA	PETEA
Proportion of multiacrylate in diluent	40%	10%	20%	30%
Bond strength (g/cm)	450	2000	1200	700

*PETEA is pentaerythritol tetra acrylate

Example 28

10 The composition of Example 18 was modified by the inclusion of 5 parts of a balsamic resin sold as ABITOL by Hercules NV of Netherlands. A sample of the formulation was then employed to fill the space between two 3 mm thick glass sheets spaced 0.75 mm apart and cured and the resultant laminate was exposed to the light of a 1 Kw high pressure mercury lamp. After 186 hours continuous exposure, the cured composition was still water-white. When the experiment was repeated using the formulation of Example 18, the cured composition turned a very pale straw colour after 186 hours exposure.

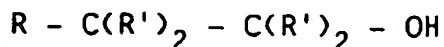
Examples 29 and 30

20 Example 28 was repeated but using 2.5 parts of balsamic resin (Example 29) and 10 parts of balsamic resin (Example 30). An improvement in aging was observed in Example 29 but the results were not quite as good as Example 28. In Example 30, the improvement over Example 28 was barely perceptible.

The claims defining the invention are as follows:

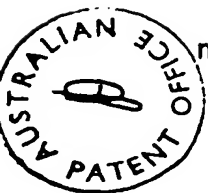
1. A radiation-curable fluid adhesive composition for the production of clear glass laminate and of the kind comprising (a) a urethane acrylate composition in (b) a liquid $\alpha\beta$ -ethylenically unsaturated diluent component comprising acrylic acid and a monoester of acrylic acid, said composition also optionally containing (c) a photoinitiator, characterised in that said urethane acrylate component comprises at least one urethane acrylate of the kind obtainable by reaction of an oligomeric urethane intermediate having end groups selected from -OH and -NCO with an $\alpha\beta$ -ethylenically unsaturated compound which has a group reactive with an end group of the intermediate and also a carboxy (-COO) group attached to a carbon atom which is attached to a methylene group by a double bond, more than 50% by weight of said urethane acrylate component is material having a molecular weight (M_N) above 1500, and said urethane acrylate component forms from 45% to 75% by weight of the composition, and said diluent component comprises (i) acrylic acid, (ii) an effective amount ^(as hereinbefore defined) of at least one acrylate selected from monoesters of acrylic acid and alkanols having from 1 to 6 carbon atoms and substituted derivatives of such alkanols, wherein each substituent contains only carbon, hydrogen and oxygen atoms, and, optionally, (iii) a multiacrylate; said component (i) being present in an amount of from 35% to 90% by weight, based on the total weight of components (i) and (ii), said component (ii) being present in an amount of from 10% to 65% by weight, based on the total weight of components (i) and (ii), and said component (iii) forming from 0% to 40% by weight of the diluent.

2. A composition as claimed in claim 1 wherein said at least one acrylate is selected from monoester of acrylic acid with alcohols having the structure



wherein R is -H, -OH or a monovalent organic group containing atoms selected only from carbon, oxygen and hydrogen, and having not more than 4 atoms in a chain attached to the free valency, and each R' is individually selected from -H and -CH₃, or R and R' together may form a divalent group containing atoms selected only from carbon, hydrogen and oxygen, the sum of the carbon and oxygen atoms in said alcohol being from 4 to 7.

3. A composition as claimed in claim 1 in which the acrylate is n-butyl acrylate.



16. A composition as claimed in any one of claims 12 to 15 in which component (b) is derived from polyol more than 50% of which is polyester polyol.

17. A composition as claimed in claim 16 in which component (b) is derived from polyol which is derived from lactone.

18. A composition as claimed in claim 17 wherein the lactone comprises caprolactone.

19. A composition as claimed in any one of claims 12 to 18 in which both component (a) and component (b) are derived from polyisocyanate wherein more than 50% of the isocyanate groups are attached to aliphatic carbon atoms.

20. A composition as claimed in any one of claims 12 to 19 wherein component (b) forms from 0.5% to 30% by weight of the total urethane acrylate oligomer.

21. A composition as claimed in any one of claims 1 to 20 wherein all or substantially all of the unsaturation in the urethane acrylate component is terminal $\alpha\beta$ -unsaturation.

22. A composition as claimed in any one of claims 1 to 21 which includes at least one further resinous material.

23. A composition as claimed in claim 22 in which said further resinous material is selected from epoxy resins and balsamic resins.

24. A composition as claimed in claim 22 or claim 23 in which said further resinous material forms not more than 50% by weight of the composition.

25. A composition as claimed in any one of claims 22 to 24 in which said further resinous material is balsamic resin and is present in an amount of up to 10% by weight of the composition.

26. A radiation-curable urethane acrylate composition as claimed in any one of claims 22 to 24 which includes an age-improving amount of a balsamic resin.

27. A composition as claimed in claim 26 in which said balsamic resin is present in an amount of from 0.5 to 50% by weight.

28. A composition as claimed in any one of claims 1 to 27 in which the acrylic acid forms at least 50% by weight of the $\alpha\beta$ -ethylenically unsaturated carboxylic acid content of the diluent.

29. A composition as claimed in any one of claims 1 to 28 in which the acrylic acid and acrylate together form at least 50% by weight of the diluent.



38. A laminate comprising a first sheet of glass bonded to a second sheet which is selected from glass sheets and clear plastics sheets by means of a layer of a composition as claimed in any one of claims 1 to 34 which has been cured by irradiation.

39. A laminate comprising a clear plastics foil or film interlayer one face of which is bonded to a glass sheet and the other face of which is bonded to a second sheet selected from glass sheets and clear plastics sheets and wherein at least the bond between the interlayer and the glass sheet is by means of a layer of a composition as claimed in any one of claims 1 to 34 which has been cured by irradiation.

40. A radiation-curable fluid adhesive composition substantially as hereinbefore described with reference to any one of the Examples.

41. A method of producing a clear glass laminate substantially as hereinbefore described with reference to any one of the Examples.

42. A laminate substantially as hereinbefore described with reference to any one of the Examples.

DATED this FOURTEENTH day of SEPTEMBER 1987

DELTAGLASS S.A.

Patent Attorneys for the Applicant
SPRUSON & FERGUSON



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